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Utilization of Catalyzed Waste Vegetable Oil as a Binder for the Production of Environmentally friendly Roofing Tiles

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ABSTRACT

Climate change has become a major issue in recent years owing to the emission of greenhouse gases. Mitigation measures are required to overcome the challenges pertained to greenhouse gases emissions. This research paper attributes to the utilization of catalyzed waste vegetable oil as a binder for the production of roofing tiles to replace the conventional construction materials such as clay and cement. A novel methodology of utilizing catalyzed waste oil incorporated with sand and filler was adopted and the innovative product produced entitled as catalyzed Vege-Roofing tiles was produced that discovered to be economical and environmentally friendly in contrast to the traditional binders. It is believed that an extended heat curing of vegetable oil results in a complex oxy-polymerization reaction converting it into a rigid binder. Triplicate prototypes

samples were manufactured to optimize the final conditions for the fabrication of catalyzed Vege-Roofing tiles. Optimized conditions were then implemented to produce standard catalyzed Vege-Roofing tiles and these fabricated tiles have shown flexural stress of up to 12 MPa for 18 hours of curing. Moreover, these novel tiles were tested for permeability and water absorption according to the ASTM standards and have shown impermeability and remarkably low water absorption. Progressively, the embodied energy and embodied carbon requirements for these tiles found to be 0.64 MJ/kg and 0.327 kg CO₂ per equivalent respectively which is quite less in comparison to the traditional binders. Conclusively, environmentally friendly and economic production of tiles, conservation of existing resources and overcoming the issue of waste management are the remarkable outcomes of this research.

Keywords: Catalyzed Vege-Roofing Tile; Catalyst; CO₂ Emission; Embodied Carbon; Embodied Energy; Waste Vegetable Oil

1. Introduction

Owing to the concern of the effects of greenhouse gases (GHGs) on our environment, it is essential to discover solutions of mitigating these gases to secure the future (Oludolapo and Charles, 2017). Surprisingly, only building industry accounts about 40% of energy utilized globally and moreover, approximately 46% of this quantity being consumed in developing countries (Hameed, 2009). Also in the course of activities involved in construction such as extraction and enormous consumption of raw materials, a huge of amount of waste is generated (Bogas et al., 2015; Angel et al., 2017). According to International Energy Agency (IEA), approximately 10% of current global man-made CO₂ emissions originated from the cement industry only (Shen et al., 2016). Furthermore, 1.84 – 2.8 kJ/kg of energy and a temperature higher than 1000 °C is required in the manufacturing of clay

43 tiles (Froth and Shaw, 2013). An enormous amount of carbon dioxide released during the
44 production of these masonry units also believed to be responsible for the enhancement of global
45 warming. This alarming situation requires being addressed seriously and environmentally friendly
46 approaches are required to be implemented at both manufacturing and extraction of building
47 materials to minimize the energy consumption.

48 Waste management is another serious issue of concern, since thousands of millions of tons of waste
49 produced per year. It is estimated that for commercial frying only in UK and US, approximately
50 50-90 million liters and 300 million gallons of used cooking oil is produced annually. Used
51 cooking oil is considered to be a waste since it pollutes the water and affect the marine life when
52 discharged into local streams (Forth and Zoorob, 2012). Utilization of waste vegetable oil in the
53 production of Biodiesel has already paved a way to overcome the issue of waste oil disposal, but
54 escalating cost of production and by-product disposal are the major hurdles in the implementation
55 of this process (Forth and Zoorob, 2012; Nadyaini et al., 2011). Similarly, fly ash that is available
56 in huge amount from thermal power plant and considered as a hazardous waste is usually land
57 filled. It is revealed that when fly ash content between 30 to 60% is used, a concrete having good
58 mechanical strength and durability could be produced (Marceau, 2002). It is also reported that a
59 20% fly ash as filler has higher compressive strength as compared to more than 50% fly ash as
60 filler (Haiying et al., 2007). Another notable consideration is that high percentage of filler could
61 have a negative impact on the compressive strength of building material (Naik et al., 2004).
62 Moreover, utilizing fly ash content greater than 50% could reduce the workability of the mixture
63 and also induce difficulty to compact the mixture (Garbacz and Sokołowska, 2013).
64 Till today, more stress is laid on investigating the replacement of the aggregates with waste
65 materials but unfortunately, a very limited focus has been paid in evaluating the energy

66 requirements of binders like cement production or kiln firing during production of concrete and
67 clay tile respectively. Cement production and firing clay bricks are considered as the foremost
68 contributors in increasing the embodied energy and CO₂ requirements (Jones and Hammond,
69 2008). Contrary, aggregate has negligible impact on the energy emissions and consequently on the
70 environment. Substituting the masonry units with renewable materials like vegetable oil having
71 comparatively lower energy and carbon emissions will definitely contribute much to overcome the
72 threatening issue of global warming. However, the concept of utilizing vegetable oil as an effective
73 building material is limited to few studies only. Oxy-polymerization reaction is considered as
74 responsible for the binding effect since it increases the viscosity and consequently hardening of
75 the vegetable oil (Quesnel, 1994; Johnson et al., 2015). It is investigated that building blocks could
76 be produced from vegetable oil mixed with recycling aggregates. These blocks have shown the
77 potential to replace the conventional building blocks (Forth and Zoorob, 2012). In addition,
78 building blocks produced by the encapsulation of vegetable oil and petroleum sludge have shown
79 high compressive strength compared to traditional building blocks (Johnson et al., 2015). It is also
80 revealed that virgin vegetable oil mixed with aggregate and filler followed by compaction and heat
81 curing could be utilized for the production of roofing tiles (Noor et al., 2015). However, energy
82 and economic constraints have limited the feasibility of these processes. Hung et al. (2015)
83 reported that a blend of waste vegetable oil and glycerol can be used in the production of masonry
84 units. The blocks produced have shown high compressive strength and low energy emissions in
85 contrast to concrete blocks. However, changes in EU Directive restricted the generation of bio-
86 fuel and consequently the glycerol's production and thus limited the feasibility of the process. It
87 was hypothesized in the present innovation that sulfuric acid (H₂SO₄) added as a catalyst to waste
88 vegetable oil would reduce the curing time and assist in developing a more energy efficient and

economical process. The present investigation aims to develop an alternate binder to replace the conventional environmentally unfriendly binders for the production of masonry units. Catalyzed Vege-Roofing tiles produced by incorporation of catalyzed waste vegetable oil and filler and sand were examined for flexural strength, water absorption, and permeability according to ASTM standards. Environmental aspects were also determined by calculating the embodied energy and carbon emissions. Furthermore, cost was also calculated for catalyzed Vege-Roofing tiles and comparative analysis was carried out with conventional concrete roofing tiles. Induction of this novel binder for the production of masonry units would expect to reduce the energy emissions and cost of the building sector to an enormous extent.

2. Experimental procedure

2.1 Materials

2.1.1 Binder

Catalyzed vegetable oil constitutes of waste vegetable oil and H_2SO_4 , was used as a binder in this investigation. Waste vegetable oil was obtained from the local restaurants in Sri Iskandar, Malaysia. Some of the properties of the waste vegetable oil examined are listed in Table 1. Moreover, 8.33 M concentrated sulfuric acid (H_2SO_4) with a percentage purity of 96 to 98% and brand name Qrec was utilized as catalyst with waste vegetable oil to reduce the curing time in the production of roofing tiles. The ratio of waste oil to H_2SO_4 used before attaining the optimized value was 25:1.

Waste vegetable oil consists of a mixture of fatty acids and mono, di- and tri-glycerides. Upon prolonged heating, some of the fatty acids may form dimers or trimers with more than one carboxylic acid group. Hence, poly-esterification can occur between di-acids and diols found in waste vegetable oil to form solid polyester and bind the tile material effectively during its curing

process. Esterification process is catalyzed by Bronsted acids, preferably by sulfonic and sulfuric acids. These catalysts give very high yields in alkyl esters (Schuchardta et al., 1998). The mechanism of the acid-catalyzed esterification between a carboxylic acid and an alcohol is shown in Fig. 1. The protonation of the carbonyl group of an acid leads to the carbonation I which, after a nucleophilic attack of the alcohol, produces the tetrahedral intermediate II, which eliminates a water molecule to form the ester and to regenerate the catalyst HA. Esterification is an equilibrium reaction and the transformation occurs essentially by mixing the reactants. However, the presence of a catalyst (typically a strong acid) accelerates considerably the rate of reaction.

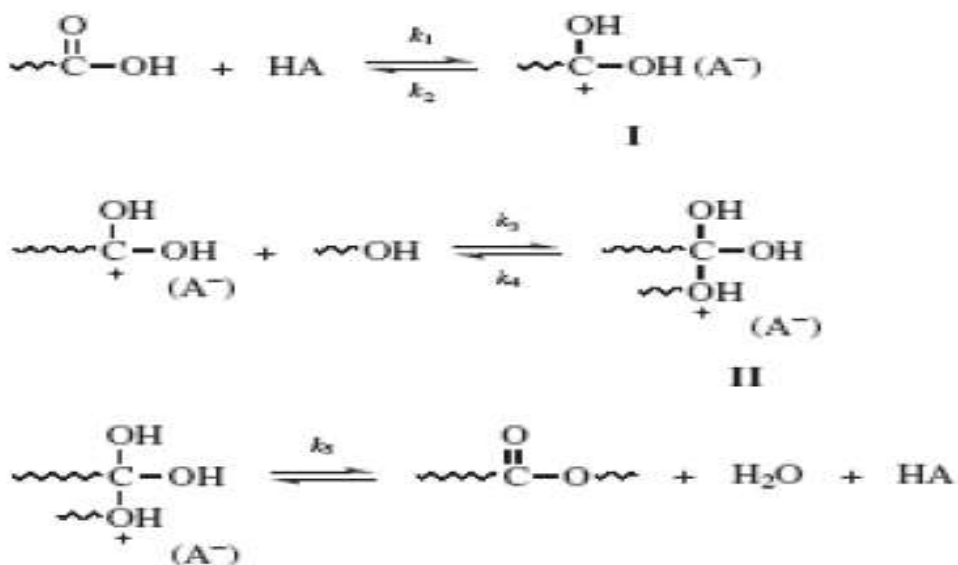


Fig.1: Mechanism of the acid-catalyzed esterification between a carboxylic acid and an alcohol

Table 1: Some tests on waste vegetable oil

Property	Used vegetable oil collected	Maximum standard limit (Berger, 2005)
Acid Value (%)	3.7	2.5
Free Fatty acids (%)	3.5	2.5

Oxidized fatty acids (%)	1	2.1
Total polar molecules (%)	30	25-27

2.1.2. Sand Aggregate

Two types of sand named as river and mining sand is used in this process. Specific gravity of both types of sands is determined by helium ultra-pycnometer and large pycnometer method following the ASTM C127-88 and C128-88. In addition, size distribution attained by sieving analysis of river and mining sand is conducted according to ASTM C 136. Size gradations of river and mining sands are presented in Fig. 2.

Table 2: Specific Gravity of Sand Types

Type	Helium Pycnometer	ASTM C127
River Sand	2.53	2.570
Mining Sand	2.67	2.647

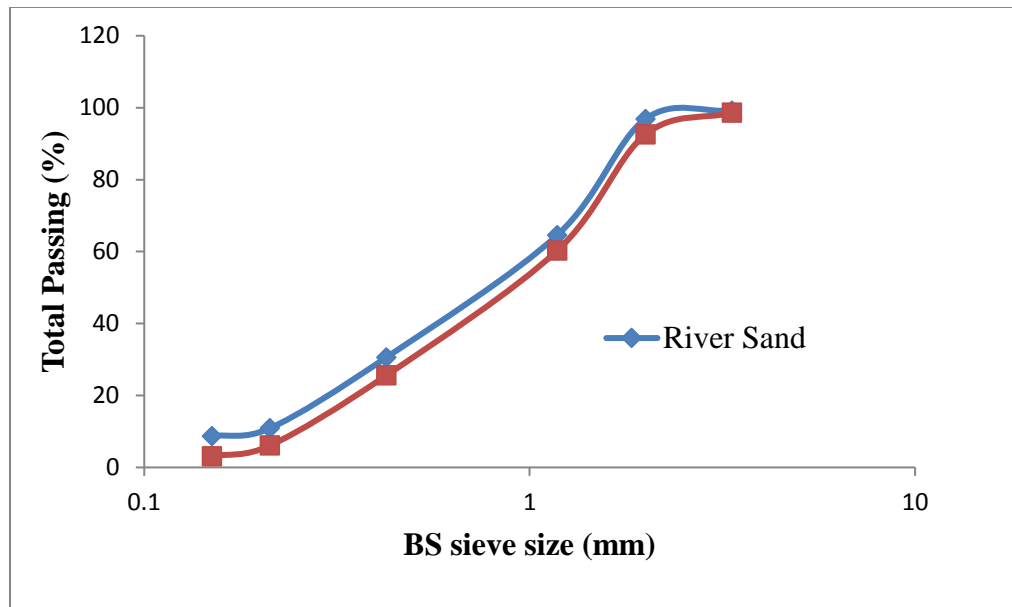


Fig. 2: Size gradation of the sand aggregates

2.1.3. Filler

Fly ash, having class F, size less than 75 μm and specific gravity of 2.5 was purchased from Kah Hwa Industries SDN. BHD, Malaysia. The typical chemical compositions and oxide analysis for fly ash used in this investigation are shown in Table 3. Fly ash was utilized as filler in the production of both prototypes samples and catalyzed Vege-Roofing tiles.

Table 3: Chemical Composition of Fly ash

Components	Mass (%)	Standard Limits (ASTM C618) %
Silicon oxide (SiO_2)	60.52	SiO_2 plus Al_2O_3 plus
Aluminium oxide (Al_2O_3)	31.12	Fe_2O_3 , min 70%
Ferrous oxide (Fe_2O_3)	1.46	Obtained 93.1%

Calcium oxide (CaO)	3.81	
Sodium oxide (Na ₂ O)	1.21	
Magnesium oxide (MgO)	0.84	
Sulfur trioxide (SO ₃)	0.73	Max 5
Chloride as Cl	0.06	
Loss of Ignition (LOI)	0.86	Max 6

143

144 2.2. Methodology

145 Primarily, parameters were required to be optimized before the fabrication of catalyzed Vege-
146 Roofing tiles. Parameters that needed to be optimized are fly ash content, curing time, the amount
147 of catalyst, blending time of catalyzed waste vegetable oil and storage life of catalyzed waste oil.
148 The temperature evaluated as appropriate for production of all samples was 190°C (Noor et al.,
149 2015). Initial appropriate values used for the optimization process of each parameter are displayed
150 in Table 4. After scrupulous mixing (catalyzed oil did not cause stickiness and gulping with
151 aggregate and filler based on physical observation) of waste oil and catalyst with aggregate and
152 filler, the mixture was transported to standard Marshall Moulds (50 mm × 100 mm) and compacted
153 with 10 blows. Triplicate prototypes samples were then heat cured in an oven maintained at a
154 temperature of 190°C and tested for flexural stress to attain the optimized values for each
155 parameter. Highest flexural stress developed was considered as a criterion for the optimization of
156 each parameter.

157

158

159 **Table 4: Initially selected values of parameters for the optimization process**

Parameter	Selected Value
Filler	35% of total (sand + fly ash) (Noor et al., 2015)
Temperature	190°C (Noor et al., 2015)
Waste oil content	8% of total (sand + fly ash). Based on physical observation, i.e it do not cause stickiness and gulping with aggregate and filler.
Acid content	6% of total waste oil used or waste oil to acid ratio of 25:1 (Based on physical observation of change of waste oil's color to dark brown)
Catalyzed Waste oil Content	Catalyzed Waste oil / Aggregate (sand) + Filler (fly ash) = 0.0945 10 minutes blending time of waste oil and catalyst Fresh catalyzed waste oil

160

161 Notably, similar ratio was chosen for catalyzed waste oil to other materials (sand and fly ash) for
 162 the production of all samples to that used for water to other materials (cement and sand) for the
 163 production of concrete roofing tiles (Johansson, 1995). However, catalyzed waste oil comprised

of both catalyst and waste oil, optimized value for the percentage of catalyst in waste oil was also needed to be calculated. Density, specific gravity, and porosity of optimized prototypes were then determined for the optimized prototypes and further tested for a percentage of water absorption and permeability respectively compiled with the ASTM standards.

After achieving the optimized values for each parameter, final catalyzed Vege-Roofing standard tiles of dimensions 390 mm x 240 mm x 10 mm were produced (Johansson, 1995). Catalyzed Vege-Roofing tiles were then examined for flexural stress, percentage of water absorbed and permeability respectively (ASTM C67-13; ASTM C 1167-03; ASTM C 1492-03; WSDOT 802). In addition, energy emissions and economic characteristics of these novel roofing tiles were also evaluated. Energy characteristics of roofing tiles were demonstrated by calculating the embodied energy and embodied carbon. Embodied energy was calculated by multiplying the amount of each material required in producing a single catalyzed Vege-Roofing tile with the embodied energy requirements of that particular material. The determination of total carbon emissions were carried out by the use of life cycle assessment (LCA) method. Environmental impact was determined for different stages of catalyzed roofing tiles such as cradle to gate, manufacturing, distribution and end of life. Carbon emission factors for processes and materials were attained by using ecoinvent 3.3. Total carbon emissions for catalyzed Vege-Roofing tiles were then assessed in accordance to LinkCycle Quick LCA tool. Assumptions that were used in calculating the embodied energy and embodied carbon are enlisted in Table 5. Cost was determined based on the raw materials and utility charges per tile and compared with the conventional concrete roofing tile. Other miscellaneous charges were excluded from the calculations. Manufacturing steps for standard catalyzed Vege- Roofing tiles are displayed in Fig. 3.

187 **Table 5:** Assumptions in calculating energy emissions

Process	Assumption
Transportation	1. Mode of Transportation was lorry 16-32 metric ton, EURO6.
	2. Waste vegetable oil was collected from local restaurants up to a distance of 1000 Km.
Production	1. Large oven having a capacity of 15 KWh was used for the production of tiles.
	2. Approximately 900 tiles were fabricated in an oven in a single batch.
Distribution	1. Roofing tiles were distributed up to a distance of 200 Km
End of Life Management	1. Used Roofing tiles were disposed of in a local site at a distance of 40 Km.

188

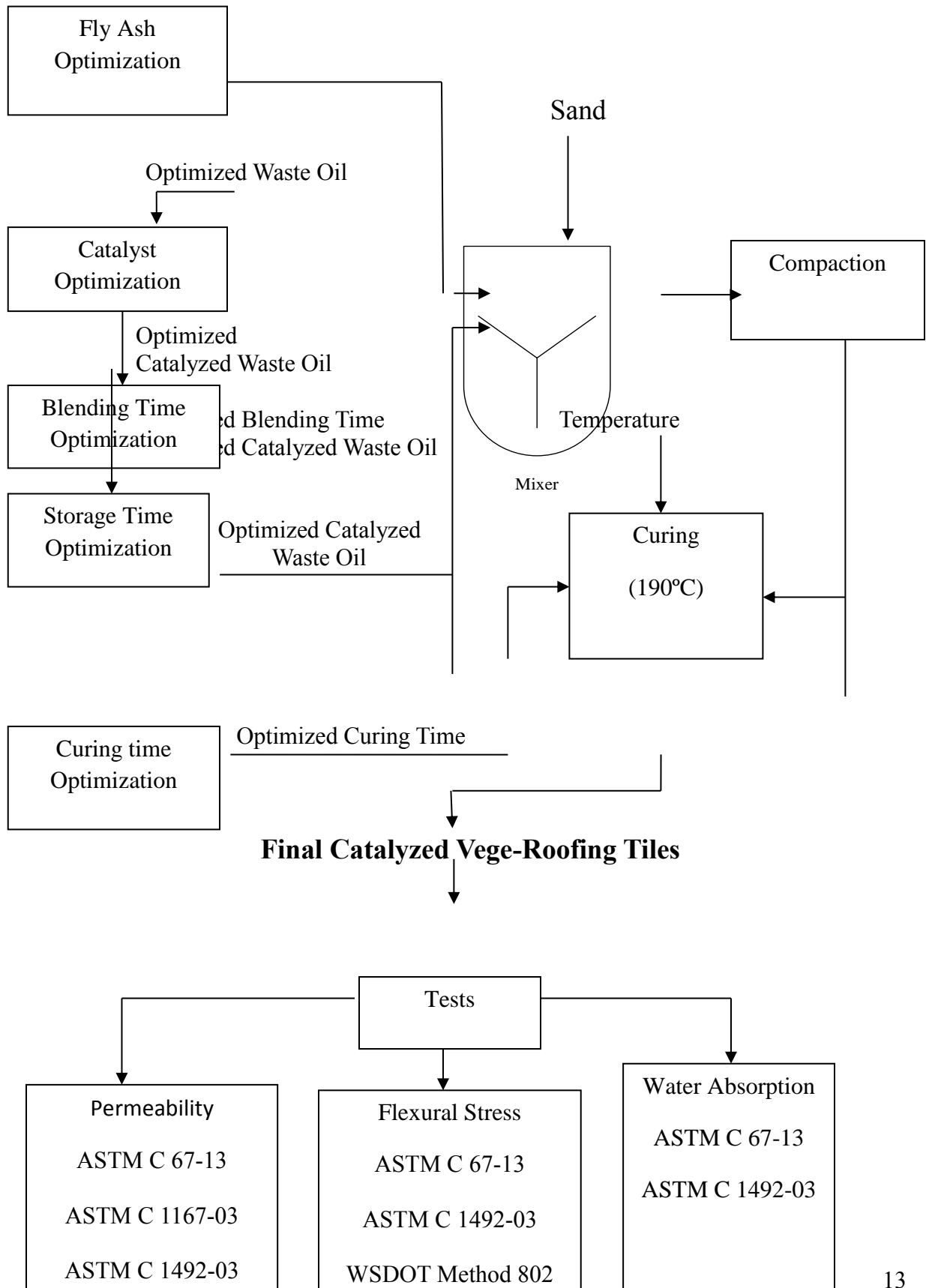
189 *2.3. Testing Procedures*

190 *2.3.1. Density and Porosity*

191 Density, specific gravity and porosity were determined for all the samples to ensure uniformity in
192 test results by using equations (1) and (2) and (3).

193
$$S.G_{mix} = (m_{filler} + m_{sand} + m_{oil} + m_{acid}) / (m_{filler}/S.G_{filler} + m_{sand}/S.G_{sand} + m_{oil}/S.G_{oil}$$

194
$$+ m_{acid}/S.G_{acid}) \quad (1)$$



213

214 **Fig. 3:** Catalyzed Vege-Roofing tile manufacturing steps

215



216

217 **Fig.4:** Catalyzed Vege-Roofing tile sample

218
$$P=100 (1-D_{mean} /(S.G)) \quad (2)$$

219
$$D= M/V \quad (3)$$

220 Density was also measured by a standard method (WSDOT TM 810) to demonstrate the validity
221 of the results.

222
$$D = A \times d_w / A-B \quad (4)$$

223 where,

224 $S.G$ =specific gravity (unit less)

225 D = density in g/cm^3

226 P = Porosity in %

227 A = Mass in grams of the surface dry sample in air

228 B = Mass in grams of the sample in water

229 d_w = density of the water at test temperature

230 2.3.2. *Water Absorption*

231 To evaluate the quantity of water that a brick can absorb, water absorption test was conducted
232 according to standard methods (ASTM C 67-13; ASTM C 1492-03).

233 The water absorption was calculated as:

$$234 \text{ Absorption } (C), \% = 100 (W_s - W_d) / W_s \quad (5)$$

235 where,

236 W_d = Mass in air in g

237 W_s = Saturated mass in water after 24 hours in g

238 Boiling water absorption can be determined as

$$239 \text{ Boiling Absorption, } B \% = 100 (W_b - W_d) / W_b \quad (6)$$

240 Finally, the saturation coefficient can be calculated as:

$$241 \text{ Saturation Coefficient} = C/B \quad (7)$$

242 2.3.3. *Permeability*

Permeability, one of the unwanted features for roofing tiles was determined for prototypes samples and standard catalyzed roofing tiles according to ASTM standard method (ASTM C 67-13; ASTM C 116703; ASTM C 1492-03). Tile's samples placed on stand in such a way that its undersides were visible and water up to approximately 10mm height allowed to sit on the top of the samples for 24 hours. The area between the samples and setup was properly sealed with a sealant to prevent the leakage. The water drops were inspected after 24 hours and if more than two of them found on the underside of the samples, then the samples would consider as significantly permeable. The experimental setup of permeability test for prototypes samples and standard tiles is shown in Fig. 5.



Fig. 5: Permeability setup for a) Prototypes samples b) Standard tiles

2.3.4. Flexural Strength

Flexural strength indicates the load that a material can withstand without breaking or rupture. Flexural strength for prototypes samples was determined by ASTM standard methods (ASTM C 67-13; ASTM C 1492-03) denoted by σ and is expressed in MPa.

258 $\sigma = MC/I$ (8)

259 where,

260 M = unit load in Newton

261 C = distance from the neutral axis in millimeters

262 I = moment of inertia in millimeters

263 Moreover, flexural stress of final roofing tiles was determined in accordance with three points
264 bending test (ASTM C 67-13; ASTM C 1492-03; WSDOT 802)

265 $\text{Flexural stress} = 3*P*L / 2*W*d^2$ (9)

266 where,

267 P =Loading force in Newton

268 L = Span Length of the tile in millimeters

269 W = Width of the tile in millimeters

270 d = Thickness of the tile in millimeters

271 **3. Results and discussions**

272 *3.1 Optimizations*

273 *3.1.1 Curing Time*

274 Triplicate prototypes specimens were produced at initially suitable conditions (35% filler, 65%
275 sand, 8% of waste oil content to aggregate and filler, waste oil to H₂SO₄ ratio of 25:1, 10 minutes

blending of catalyzed waste oil) and cured for different hours at a temperature of 190°C. Fig. 6 presents the trend of flexural stress developed for varying curing time at a temperature of 190°C. It is revealed from Fig. 6 that the flexural stress discovered to be highest for 16 and 20 hours of curing. Moreover, after 20 hours of curing at a temperature of 190°C, flexural stress began to decline to an appreciable extent probably due to the reason that prolonged curing induced the cracks internally causing the strength of prototypes specimens to reduce. It is also discovered from Fig.6 that flexural stress achieved for all days of curing has fulfilled the standard minimum requirement of 6 MPa (Crow, 2000). Moreover, standard deviation for each value of flexural stress found to be less than 0.01 thus indicating a well-controlled process (NRMCA, 2000). Efficient curing of the tiles is extremely important since insufficient curing can reduce the strength to a tremendous extent. To ensure the adequate curing of the final tiles and taking into account the energy perspective, the average of two curing times that showed highest flexural stress i.e. 18 hours of curing considered as an optimal curing time for the production.

3.1.2 Filler Content

To acquire suitable filler percentage, triplicate prototype specimens were produced at initially suitable conditions of catalyzed waste oil and optimized curing time of 18 hours and filler percentages were varied between 30% and 50%. Filler content was restrained between 30 and 50% since utilizing fly ash content greater than 50% could reduce the workability of the mixture and also induce difficulty to compact the mixture (Garbacz and Sokołowska, 2013). Flexural stress of triplicate prototype specimens at varying filler percentages was determined and illustrated in Fig. 7.

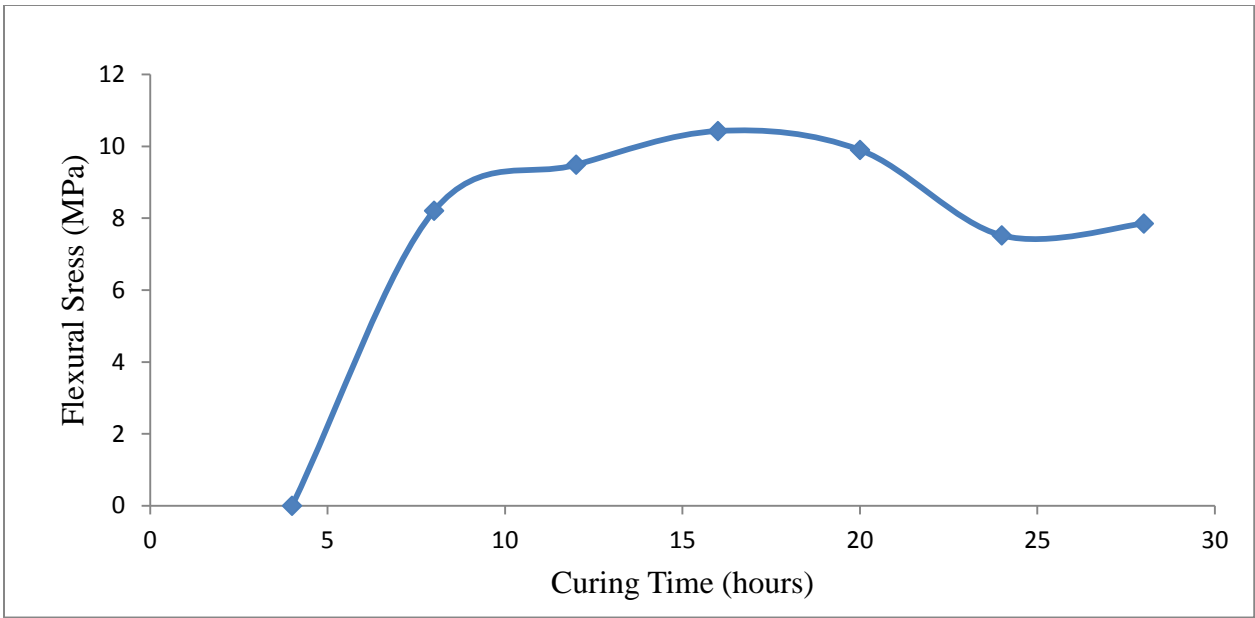


Fig. 6: Optimization of curing time

The trend of Fig. 7 reveals that flexural stress was on the lower side with 30% fly ash as filler. By increasing the fly ash content to 35%, highest flexural stress of approximately 9 MPa was achieved. However, further addition of fly ash reduced the flexural stress for the samples as observed for filler percentages of 40%, 45% and 50% respectively. However, no specific mechanism is available for the reaction of vegetable oil and fly ash but it is observed that fly ash improves the binding characteristics of vegetable oil and enhances the flexural and tensile properties due to the presence of high silica and alumina content (Micheal et al., 2014; Saumya et al., 2016). Moreover, fly ash has a neutralizing effect in strong acids such as sulfuric acid i.e. when introduced into strong acids it tends to lower the pH (Manisha et al., 2009). This is a remarkable effect since it is an indication that catalyzed Vege-Roofing tiles when exposed to fire would be non-flammable. Nonetheless, the flexural stress developed and standard deviations for each filler percentage were higher in contrast to standard minimum requirement (CROW, 2000; NRMCA, 2000). It can be

deduced from Fig. 7 that 35% of fly ash as filler showed highest flexural stress and considered as the optimal percentage of filler content.

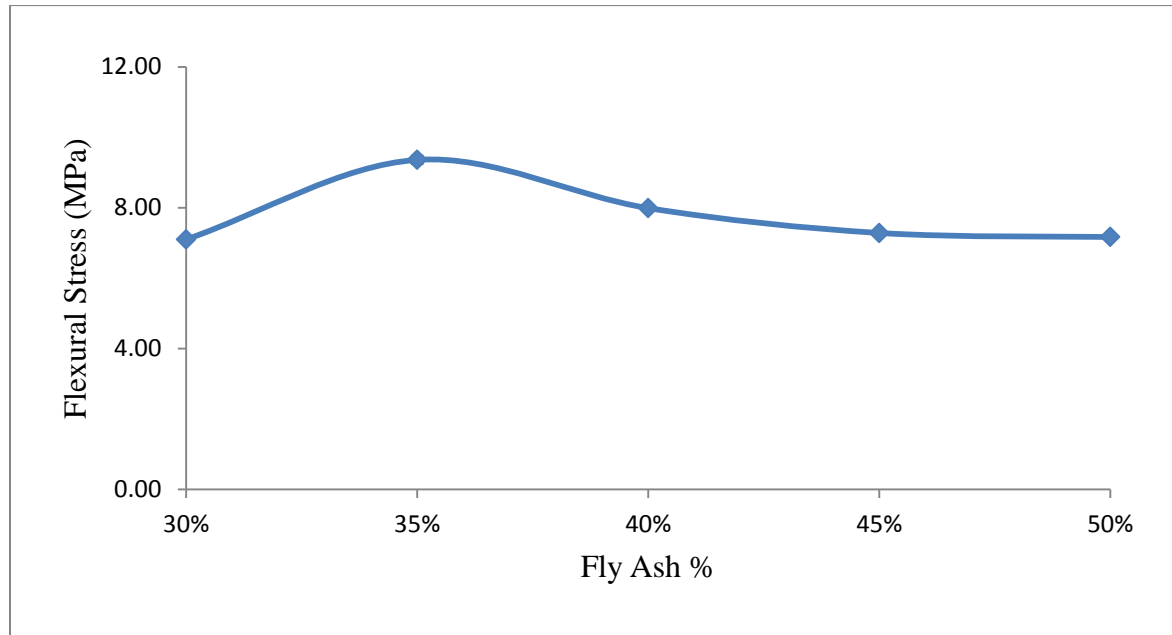


Fig. 7: Optimization of filler content

3.1.3 Percentage of Acid

Triplicate prototypes samples were produced at initially selected conditions of blending and storage time and optimized fly ash content of 35% and optimized curing time of 18 hours. Acid percentage to waste oil is altered in the range of 3% to 18 %. The range was chosen based on physical observation since introducing 3% of acid into waste oil just started the color change of catalyzed waste oil blend. Moreover, increasing percentage of an acid in oil beyond 18% reduced the workability of the mixture since the addition of an acid into waste oil tends to increase the viscosity the mixture of acid and oil. Fig. 8 exhibits the flexural stress achieved with varying percentage of acid to waste oil. It is demonstrated from Fig. 8 that flexural stress found to be

highest between 3 to 6 percent of acid in waste oil. It is also discovered that increasing the percentage of acid in waste oil reduced the flexural stress of samples. This is probably due to the increased viscosity of the catalyzed mixture when additional acid added into waste oil. Oxy-polymerization reaction is considered responsible for increased viscosity of the oil (Quesnel, 1994; Johnson et al., 2015). It is revealed that upon prolonged heating, some of the fatty acids in waste vegetable may form dimers or trimers with more than one carboxylic acid group. Hence, polyesterification can occur between di-acids and diols found in waste vegetable oil to form solid polyester and bind the tile material effectively during its curing process at 190°C. It is also believed the presence of a catalyst (typically a strong acid like H₂SO₄) accelerates considerably the rate of reaction and effectively reduced the curing time to about 18 hours (Schuchardta et al., 1998). The flexural stress and standard deviation achieved for each acid percentage calculated to be 8 to 10 MPa and 0.001 to 0.4 MPa respectively which is well within the practical standard limits (CROW, 2000; NRMCA, 2000). Highest flexural stress was achieved for 3 to 4 percent of acid in waste oil and was utilized for fabrication of standard catalyzed Vege-Roofing tiles.

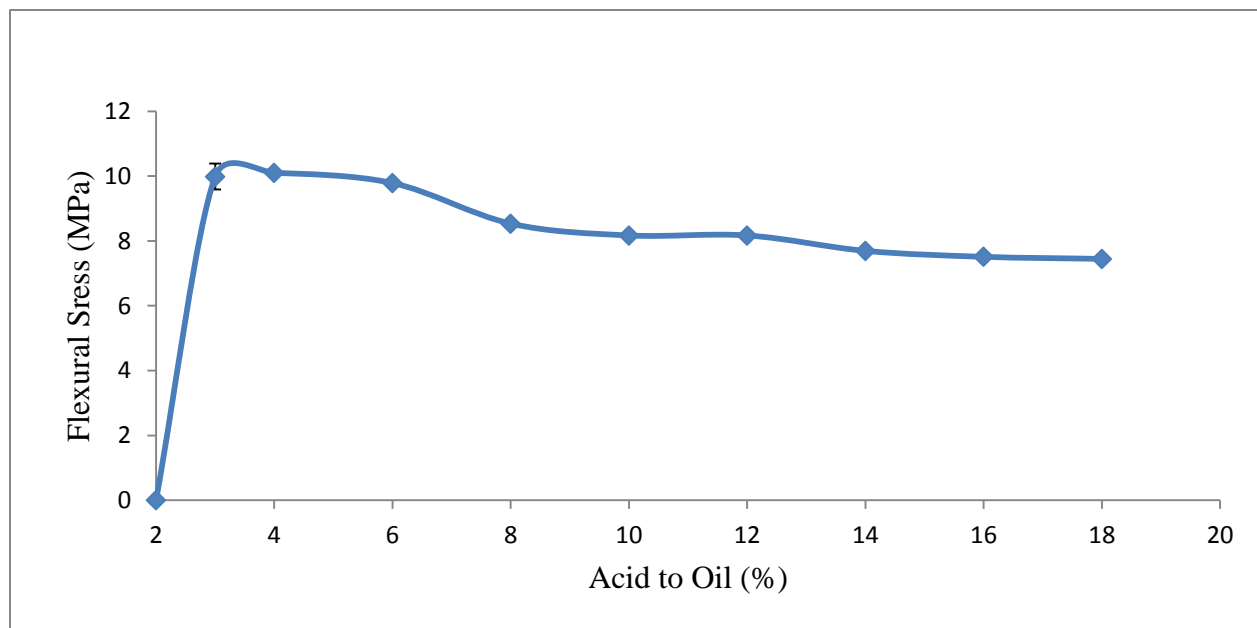


Fig. 8: Optimization of Acid content

3.1.4. Blending Time for Catalyzed Waste Oil

Blending or mixing time of catalyzed waste oil (mixture of acid and waste oil) is considered as one of the significant parameters to be reported since proper mixing of waste oil and acid could improve the mechanical properties of the catalyzed Vege-Roofing tiles. Waste oil and acid were blended for different times and triplicate prototypes samples were produced from each blend utilized as fresh at optimized conditions of filler, curing time and percentage of acid in waste oil. Blending time for catalyzed waste oil was then optimized by demonstrating the flexural stress of triplicate prototypes samples produced. Fig. 9 presents the flexural stress achieved by prototypes samples produced from various blending times of optimized catalyzed waste oil at 190°C. It is illustrated by Fig. 9, that flexural stress developed found to be highest for blending times of 10, 20 and 40 minutes respectively. Interestingly, after 40 minutes of blending the flexural stress did not change much probably due the reason that optimum mixing has been achieved earlier. However, from energy perspective, 10 minutes of blending was evaluated as an optimal blending time for the production of standard catalyzed Vege-Roofing tiles.

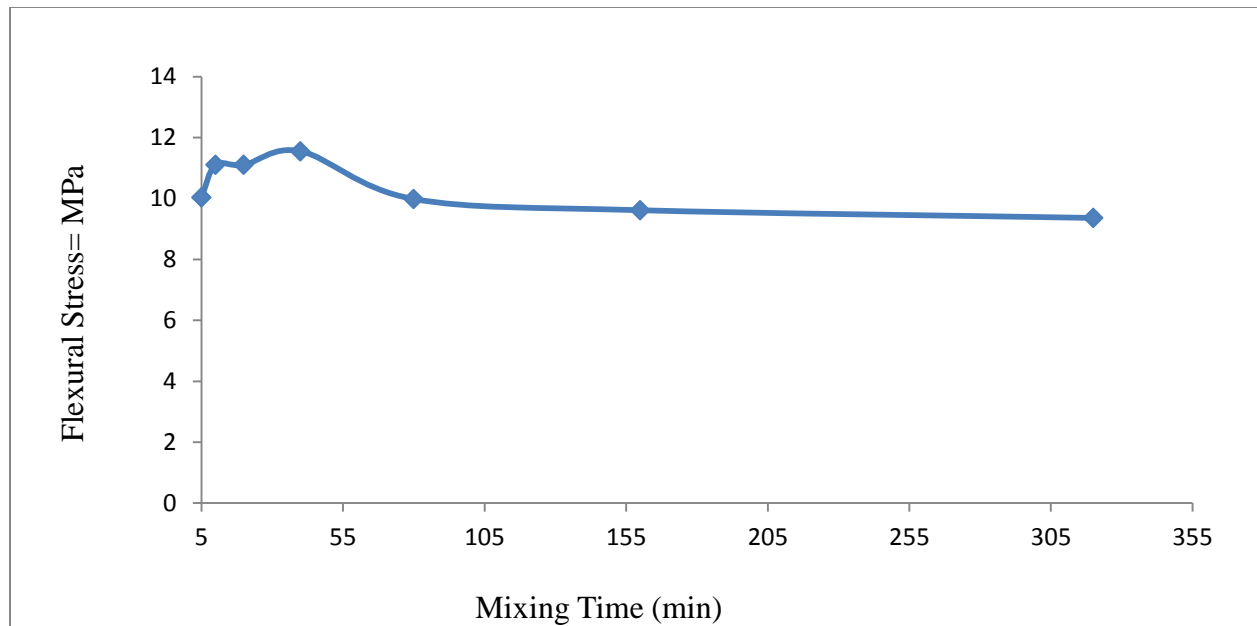


Fig.9: Optimization of blending time of catalyzed waste oil

3.1.5 Storage Life for Catalyzed Waste Oil

Storage life is an indication of storing the material while remaining within a safe limit. Storage life consideration of catalyzed waste oil is important especially from industrial point of view. Flexural stress for triplicate prototypes samples produced from fresh and stored catalyzed waste oil at already optimized conditions is displayed in Fig. 10. Notably, after 3 days of storage, catalyzed waste oil evaluated as no longer workable with the aggregate and filler since it converted to a very rigid material. It is revealed from the Fig. 8 that flexural stress found to be on the higher side for fresh catalyzed waste oil and started to decline with days of storage. It is shown in Fig.10 that utilizing the freshly catalyzed waste oil attained a flexural stress of approximately 11 MPa which is quite high in contrast to the standard minimum requirement of 6 MPa (CROW, 2000). Additionally, the standard deviation of flexural stress for each day of storage of catalyzed waste

oil found to be less than 0.1 MPa. Thus freshly catalyzed waste vegetable oil was evaluated as the optimal value for the fabrication of standard catalyzed Vege-Roofing tiles.

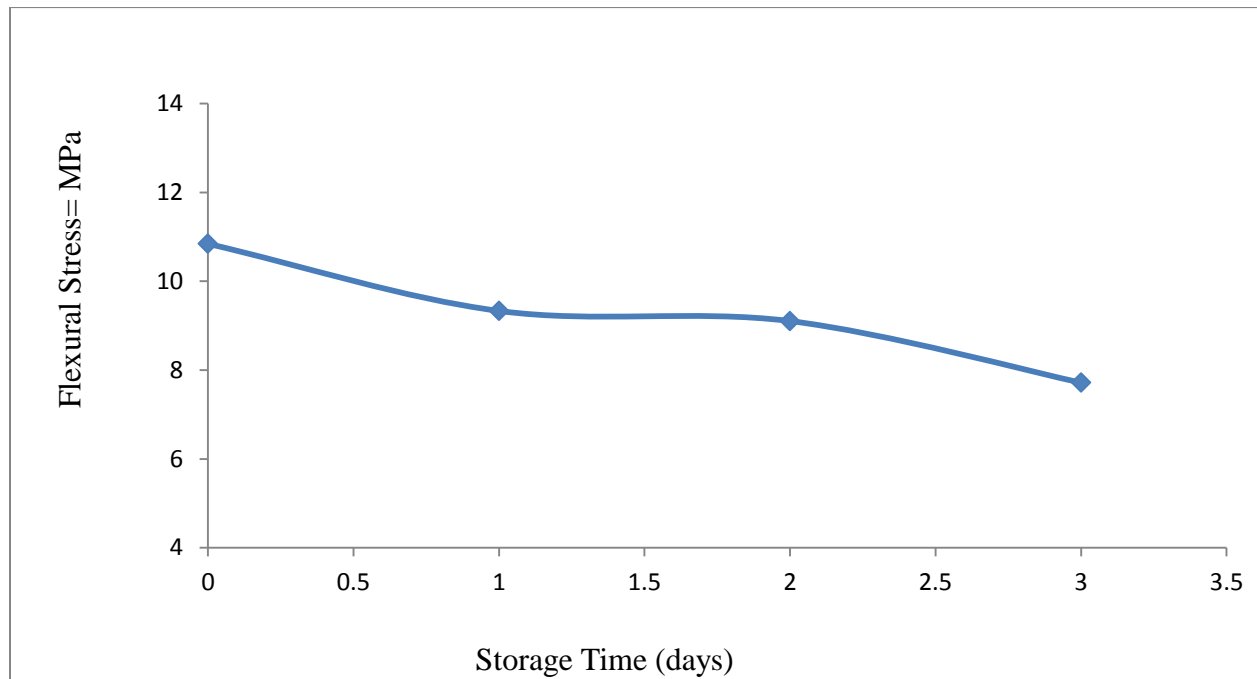


Fig. 10: Storage life optimization of catalyzed waste Oil

3.2 Optimized Prototypes

Prototypes samples were produced with already optimized parameter and examined before the fabrication of standard catalyzed Vege-Roofing tiles. Density, porosity, permeability and percentage of water absorption were calculated for optimized prototypes samples. Table 6 shows that high density specimens indicated low porosity while specimens that had low density pointed relatively high porosity. This is an indication that a dense material has fewer chances to leak or have low porosity. Results of Table 6 also reveal that percentage of water absorption for prototypes samples produced was in the range of 1.8% to 2.6% which is quite less in contrast to the standard

practical limit (Johansson, 1995; ASTM C 1492-03). Moreover, it should also be taken into account that all the optimized prototypes samples have passed the permeability test. The low percentage of water absorption is a reflection of the low porosity of the tiles (Hung et al., 2015; Zhang and Zong, 2014). Moreover, higher the porosity more will be the probability that the material becomes permeable (Ekstorm, 2001). Generally, greater the permeability, lower will be the durability performance of concrete (Khan and Lynsdale, 2002). Less porosity, low percentage of water absorption and impermeability enhance the durability of the tiles (Farhana et al., 2015).

Table 6: Density and porosity of optimized prototypes specimens (35% filler, 65% sand, 8% oil, 0.5% acid, $S.G_{\text{filler}}=2.5$, $S.G_{\text{sand}}=2.66$, $S.G_{\text{oil}} = 0.84$, $S.G_{\text{acid}}=1.83$, $S.G_{\text{mix}}=2.1$)

No.	Mass (g)	Density g/cm ³	Porosity (%)	Water absorption (%)	Permeability Test
1	164.0	2.08	1.0	2.12	PASS
2	162.5	2.06	2.0	2.45	PASS
3	163.5	2.07	1.5	1.98	PASS
4	162.1	2.05	2.4	2.76	PASS
5	160.9	2.04	2.9	3.16	PASS

3.3 Standard Catalyzed Vege-Roofing tiles

Optimized prototypes samples found to be impermeable and showed a low percentage of water absorption. Thus, the utilization of catalyzed vegetable oil was further investigated by producing standard catalyzed Vege-Roofing tiles at already optimized conditions at a temperature of 190°C. Initially, density and porosity of standard catalyzed Vege-Roofing tiles were calculated and indicated in Table 7. The bulk density was approximately 1.98 to 2 g/cm³ while the density varies from 1.7 to 2.4 g/cm³ for lightweight to normal concrete (Richard, 2004). It is found that denser materials have fewer chances to leak (Dias, 2000). It is also revealed from the Table 7 that denser materials had low porosity and vice versa (Dias, 2000 ; Ekstorm, 2001).

Table 7: Density and porosity of standard catalyzed Vege-Roofing tiles cured for 18 hours (35% filler, 65% sand, 8% oil, 0.5% acid, S.G_{filler}=2.5, S.G_{sand}=2.66, S.G_{acid}=1.83, S.G_{mix}=2.1)

No.	Mass (g)	Density, M/V g/cm ³	Density WSDOT TM 810 g/cm ³	Density (Mean)	Porosity (%)
1	1870	1.99	1.97	1.98	5.8
2	1882	2.00	1.99	1.99	5.3
3	1904	2.03	2.00	2.01	4.3
4	1849	1.97	1.98	1.98	5.8
5	1858	1.98	2.00	1.99	5.3

3.3.1 Flexural Stress & Breaking Strength

Flexural stress and breaking strength calculated for catalyzed Vege-Roofing tiles produced at optimized parameters are displayed in Table 8. It can be seen from Table 8 that flexural stress developed for standard catalyzed Vege-Roofing tiles found to be in the range of 11.4 to 12.2 MPa respectively. This suggests that the average flexural stress achieved by catalyzed Vege-Roofing tiles was approximately twice in contrast to the minimum standard requirements of 6 MPa (CROW, 2000). Nonetheless, the values of flexural stress obtained were much higher as compared to the BS 6073. This is due to an excellent binding ability of waste vegetable oil incorporated with fly ash and sand. Heat curing of waste vegetable oil initiated the oxy-polymerization reaction and converted it into a solid rigid binder (Johnson, 2015; Quesnel, 2009). Thus, a high flexural stress was developed with waste vegetable oil in comparison to other wastes such as cotton and limestone powder wastes that achieved a maximum flexural stress up to 3.5 MPa (Halil and Tugut, 2007). Also, breaking strength for concrete roofing tiles should be at least 550 to 600 N and contrary, breaking strength achieved by catalyzed Vege-Roofing tiles was higher in comparison to minimum breaking strength requirements of concrete roofing tiles (Wood and Hack, 1986; BS EN, 2011). Moreover, the standard deviation for the five tiles tested for flexural stress was below 0.7 MPa indicating a well controlled process (NRMCA, 2000). This suggests that catalyzed Vege-Roofing tiles have fulfilled the minimum standard requirements of flexural stress for the production of roofing tiles.

Table 8: Breaking strength and flexural stress of optimized standard catalyzed Vege-Roofing tiles cured for 18 hours at 190°C (Span Length=130mm, Width of tiles=240mm, Depth of tiles=10mm)

Tile No.	Loading force, P (N)	Breaking Strength (N)	Flexural Stress (MPa)
1	1400	758.3	11.4
2	1500	812.5	12.2
3	1500	812.5	12.2
4	1500	812.5	12.2
5	1400	758.3	11.4

429

430 3.3.2 Water Absorption

431 Percentage of water absorption for standard catalyzed Vege-Roofing tiles calculated according to
432 standard method and the results are demonstrated in Table 9. Table 9 indicates that percentage of
433 water absorption for five catalyzed Vege-Roofing tiles was found to be in the range of 4.7 to 5.4
434 percent. It indicates that values of water absorption attained were within the standard limit
435 (Johansson, 1995; ASTM C 1492-03; Donald and Grail, 1985). The low percentage of water
436 absorption was thought to be due to the low porosity of the tiles (Hung et al., 2015; Zhang and
437 Zong, 2014). In addition, the boiling water absorption is usually more as compared to fresh water
438 absorption. The average boiling absorption was approximately 7.4% which is low in contrast to

the boiling water absorption of building blocks produced from encapsulation vegetable oil and petroleum sludge (Johnson et al., 2015). Since the percentage of water absorption under boiling is still within the standard practical limit, it indicates that catalyzed Vege-Roofing tiles can work well in extremely hot and humid conditions (ASTM C67-13; ASTM C1492-03). Boiling water absorption is usually determined to find the saturation coefficient of tiles. Saturation coefficient of catalyzed Vege-Roofing tiles found to be in the range of 0.64 to 0.78 which is within the standard limit of less than 1 (BIA, 2007). A low value of saturation coefficient is an indication that the tiles are less absorptive and more durable while tiles with high saturation coefficients are susceptible to damage (Abdullah et al., 2015). Furthermore, all the catalyzed Vege-Roofing tiles tested for permeability have passed the test. Less porous materials are denser and denser materials have fewer chances to leak and vice versa. Thus higher the density lower will be the porosity and permeability (Dias, 2000; Zhang and Zong, 2014). Low porosity, low percentage of water absorption and impermeability increases the durability of the tiles (Farhana et al., 2015).

3.3.3 Energy Characteristics

Embodied energy and embodied carbon for producing catalyzed Vege-Roofing tiles were assessed to ensure the environmental suitability of the product. Embodied energy for same batch of tiles produced per catalyzed Vege-Roofing tiles is displayed in Table 10. Since fly ash is the by-product produced during combustion of coal in electricity generation, it has no process energy. In addition, embodied energy of fly ash is zero since it is a waste and its collection is obligatory (Chani et al., 2003; Ostwal and Chitawadagi, 2014).

Table 9: Percentage of water absorption and saturation coefficient for optimized standard catalyzed Vege-Roofing tiles

No.	Dry Weight of Tile, W_d	Saturated Weight of Tile, W_s	Absorption % (Cold Water)	Saturated weight of tile after 5 hr in boiling water, W_b	Absorption % (Boiling Water)	Saturation Coefficient
1	1970.5	2072.5	5.2	2110.2	7.1	0.73
2	1982.2	2076.5	4.7	2145.6	7.7	0.61
3	1968.3	2070.0	5.2	2128.1	8.1	0.64
4	2012.5	2107.4	4.7	2153.4	7.0	0.67
5	2002.6	2110.5	5.4	2141.5	6.9	0.78

Table 10: Embodied energy requirements in one catalyzed Vege-Roofing tile

Material	Embodied Energy (MJ/kg)	Material Required per tile (kg, L)	Total Embodied Energy per tile (MJ/kg)
Sulfuric Acid	5.00 ^[a]	0.006	0.03
Waste Vegetable oil	2.00 ^[b]	0.157	0.31
Processing	0.06 ^[c]	-	0.06
Sand	0.20 ^[d]	1.128	0.23
Fly Ash	0.00 ^[e]	0.608	0
			0.64

*a, (Eric et al, 2002); b, (Reijnders and Huijbregts, 2008); c, (Francois, 2001); d, (Ecoinvent 3.3, 2016); e, (Chani et al, 2003; Ostwal and Chitawadagi, 2014).

It is discovered from Table 10 that the embodied energy per catalyzed Vege-Roofing tile found to be 0.64 MJ/kg. This indicates that the embodied energy requirement in producing single catalyzed Vege-Roofing tiles was quite low in comparison to the embodied energy required to produce conventional clay and concrete roofing tiles (Hammond and Jones, 2008). Comparative analysis of embodied energy for similar dimensions of conventional roofing tiles and catalyzed Vege-Roofing tile is presented in Table 11. It is discovered from Table 11 that the embodied energy of catalyzed Vege-Roofing tile is 321% less than conventional concrete tile, 837% less than clay tile and 1775% less than ceramic tile as calculated by Hammond and Jones (2008). Embodied carbon in producing one catalyzed Vege-Roofing tile for different phases is demonstrated in Table 12.

Table 11: Comparative analysis of embodied energy

S. No	Tile (390mmx240mmx10mm)	Total Embodied Energy (MJ)
1	Concrete	2.7
2	Clay	6.0
3	Ceramic	12.0
4	Catalyzed Vege	0.64

480

481 **Table 12:** *Total carbon emissions in different phases of catalyzed Vege-Roofing tile

Cradle to Gate Emissions					
Material	Quantity (kg)	Emission factor kg CO ₂ /equiv.	Transport (Km)	Transport	Total Emission
				Emissions (per kg, Km)	
Waste Oil	0.1574	0	1000	0.0001	0.100
Sulfuric Acid	0.0066	0.17	40	0.0001	0.005
Fly Ash	0.6080	0.004	40	0.0001	0.006
Sand	1.1280	0	20	0.0001	0.002
Total Phase Emission					0.113
Manufacturing					
Operation	Curing Time (hours)	Electricity Usage (KWh)	Emission Factor (kg CO2 per equiv.)		Total Phase Emission
Heat Curing	18	0.3	0.63		0.19
Distribution					

Material	Transport (Km)	Transport Emissions (per kg, Km)	Total Phase Emission
Roofing Tile	200	0.0001	0.020
End of Life			
Material	Transport (Km)	Transport Emissions (per kg, Km)	Total Phase Emission
Roofing Tile	40	0.0001	0.004
Total			0.327

*Emission factors values were demonstrated from Ecoinvent 3.3.

Table 12 reveals that estimated CO₂ emission value per tile for the same batch of tiles was calculated to be 0.327 kg CO₂/equivalent, which is low in contrast to the traditional roofing tiles. The carbon emissions for catalyzed Vege-Roofing tiles were 202%, 49% and 205% lower in contrast to concrete, clay and ceramic roofing tiles as determined by ecoinvent 3.3. Moreover, the emission values determined for novel tiles were 267%, 37% and 126% lower than concrete, clay and ceramic tiles as estimated by Hammond and Jones, (2008). This indicates that catalyzed Vege-Roofing tiles if implemented would be environmentally friendly and will reduce the energy emissions to an appreciable amount. Comparison of energy emission values for catalyzed Vege-Roofing tiles and conventional roofing tiles is displayed in Fig. 11.

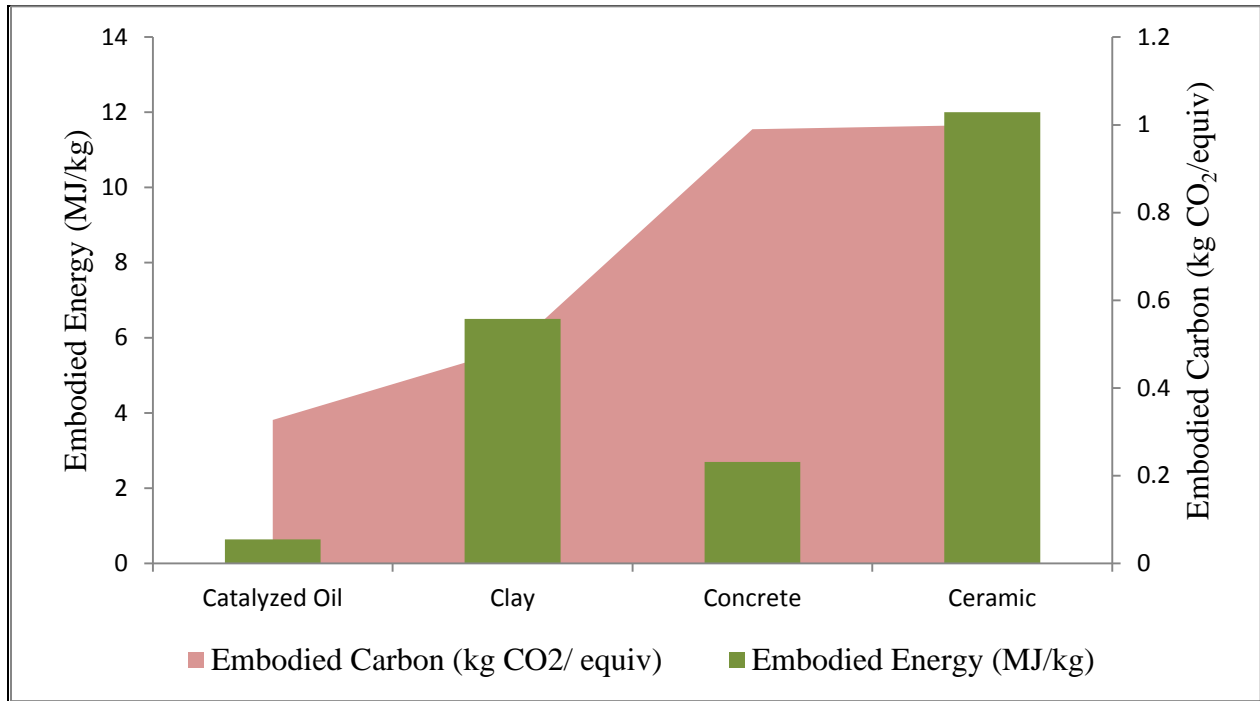


Fig. 11: Energy Comparison of catalyzed and traditional roofing tiles

Fig. 11 indicates that catalyzed Vege-Roofing tiles are more environmentally friendly since the energy emissions discovered to be on the lower side in contrast to traditional roofing tiles. Low energy emissions are due to the replacement of masonry units such as kiln firing in clay and cement production (Hammond and Jones, 2008). Traditional high energy consuming binders were replaced by waste binder i.e. waste vegetable oil that reduced the energy emissions to a remarkable extent.

3.3.4 Economic Evaluation

The economy of the product is also evaluated as one of the important criteria to determine the feasibility of the product. Cost of power and water consumption for industrial sector found to be around RM 0.38 per KWh and RM 2.07 per cubic meter as retrieved from Tenaga Nasional Berhad

and Syarikat Bekalan Air Selangor SDN. BHD, Malaysia respectively. Fly ash, sand, cement, sulfuric acid and waste palm oil could be purchased from local suppliers in Malaysia at a price of RM 80, RM 80, RM 400, RM 500 and RM 400 per metric ton respectively. For cemented tile, cost was determined by using the cement to sand ratio of 1:4 (Johansson, 1995) mixed with an appropriate quantity of water. However, additional amount of water needed for the hardening process of these cemented tiles was also estimated for cost calculation. Cost comparison of same batch of catalyzed Vege-Roofing tiles and concrete roofing tiles is displayed in Table 13. Rate per tile was calculated by multiplying the material used in producing one tile with the rate of that particular material. Table 13 shows that the cost of catalyzed Vege-Roofing tile was comparatively less in contrast to conventional concrete roofing tile. Utilization of catalyst induced a major contribution to reduce the cost of the process since it reduced the curing time of the production.

526 **Table 13:** Economic comparison of cemented and catalyzed Vege-Roofing tiles (a) Raw
527 material/tile (b) Utilities/tile

528

5 **a)**

Cemented Tiles

Catalyzed Vege-Roofing tiles

S.No.	Description	Rate (RM)	Rate per tile (RM)	S.No.	Description	Rate (RM)	Rate per Tile (RM)
1	Cement	400/metric ton	0.170	1	Fly ash	80/metric ton	0.047
2	Sand	80/metric ton	0.102	2	Sand	80/metric ton	0.077
				3	Waste Oil	400/metric ton	0.034
				4	Sulfuric Acid	500/metric ton	0.003

b)

1	Power Cost	0.38/KWh	0.06	1	Oven	0.38/KWh	0.12
2	Water	2.07/cubic meter	Approx 0.05	2	Mixer	0.38/KWh	0.001
		TOTAL	0.382			TOTAL	0.282

530 **4. Conclusions**

This research attributes to the production of catalyzed Vege Roofing tiles and discovered that catalyzed waste vegetable oil can be used as an alternate binder for the manufacturing of roofing tiles. The fabricated catalyzed Vege Roofing tiles has met the criteria for standard concrete or kiln burned clay tile as it indicated high flexural strength, low water absorption and impermeability. Implementation of novel roofing tiles would be economical and energy efficient process since energy requirements and cost are comparatively low in contrast to traditional roofing tiles. Production of this novel bio-composite also paved a way for the production of other building materials such as building blocks, flooring etc. by industrial symbiosis and thus converting the building production to a more cleaner and greener process. Additionally, this research has a scope to eliminate waste disposal problems, since fly ash, a waste from thermal power plant and waste vegetable oil, a waste from local restaurants being utilized in producing these novel tiles. **However, due to the formation of solid polyester as a binder in this work, UV degradation should be considered as a special care to use the catalyzed Vege-tile for roofing.** This novel process for the production of building materials would help in conserving the existing resources. Environmentally friendly production of building materials, low cost production and the waste management are the remarkable outcomes of this research.

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Nomenclature

ASTM	American Society for Testing and Materials
BHD	Berhad (Private)
MPa	mega-pascal (unit of strength)
MJ/kg	Mega-joule per kilogram (unit of energy)

KWh	Kilowatt hour (unit of power)
RM	Malaysian Ringgit (currency)
WSDOT	Washington State Department of Transportation
